X-Ray Identification of Element 104*

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The daughter x-ray identification technique has been applied to the identification of element 104. The characteristic K-series x rays from the α-decay daughter isotope, nobelium ($Z=102$), have been observed in coincidence with α particles from the decay of 4.5-sec $^{257}104$, thus providing an unequivocal determination of the parent atomic number, $Z=104$.

Many isotopes of the transfermium elements ($Z>100$) have been synthesized in recent years and their elemental or atomic number assignments have usually been based on a combination of transmutation data and, in a few cases, on chemical properties. The transmutation data include α and spontaneous-fission decay systematics, genetic-linkage experiments, cross-section systematics, excitation-function measurements, and cross bombardments with different projectiles and/or targets. The best evidence for the identification of a new element, however, is the observation of characteristic K- or L-series x rays,[1,2] as first utilized by Moseley[3] in 1914, since the energies of these x rays are directly connected to the atomic number of the element.

Although Moseley[3] and Barton, Robinson, and Perlman[4] could rely on a simple relationship between the atomic number and the corresponding x-ray energies, a somewhat different technique must be used to predict the energies of K-series x rays for the heaviest elements ($Z>95$). These energies have been calculated by Carlson *et al.*[5] using the Oak Ridge relativistic Hartree-Fock-Slater computer program. The x-ray–energy and electron-binding–energy predictions of Carlson *et al.*[5] have been tested experimentally through $Z=100$ and have been shown to be accurate to within ±30 eV (or about 0.02%).[6-8] In addition, the intensities of K-series x rays for the heaviest elements have been calculated in the relativistic treatments[9,10] and have also been verified experimentally.[7] Thus, even for the very heavy elements, the measurement of the characteristic K-series x rays provides a firm basis for a direct and unequivocal atomic-number assignment.

Recently, we reported the conclusive determination of the atomic number of a nobelium isotope ($Z=102$),[2] using a modified x-ray identification technique. This technique relies on the observation of x rays from the daughter element in coincidence with the α particles from the decay of the parent element. These x rays can arise from atomic rearrangements following internal conversion processes in the de-excitation of the daughter element, if the α decay should proceed to excited nuclear states. We would like to report in this paper the application of this modified x-ray identification technique to the conclusive identification of element 104.

The 4.5-sec, α-emitting isotope $^{257}104$, first produced in 1969 by Ghiorso and co-workers,[11-13] was produced at the Oak Ridge isochronous cyclotron laboratory in the reaction $^{249}\mathrm{Cf}(^{12}\mathrm{C}, 4n)^{257}104$. The target consisted of 220 μg of isotopically pure ^{249}Cf which had been electrodeposited on a 2.5-mg/cm^2 Be foil over an area of 0.36 cm^2. Our measured cross section for the production of $^{257}104$ was $\sim 1.2\times10^{-32}$ cm^2 at an energy of 73.0 MeV.

The reaction products, recoiling out of the ^{249}Cf target, were thermalized in a small helium-filled chamber, continually pumped through a 0.013-in. orifice, and collected on a small disk of aluminum (1.43 cm diam and 0.053 cm thick). After a bombardment and collection time of about 10 sec, the catcher disk was pneumatically transferred a distance of 10 m in 1.7 sec to a counting station located outside the heavily shielded bombardment room. The transferred disk was automatically positioned between an α-particle detector and a high-resolution photon detector.

The α-particle detector was a 200-mm^2 Si(Au) surface-barrier detector which subtended a solid angle of 38.5% of 4π sr. The measured energy resolution for α particles was $\sim$30 keV full width at half-maximum (FWHM) under our experimental conditions. The photon detector was a high-resolution Ge(Li) planar detector (25 mm diam with 7 mm depletion depth) which was equipped with a 24-mg/cm^2 Be entrance window. The energy resolution for 122-keV γ rays (comparable in

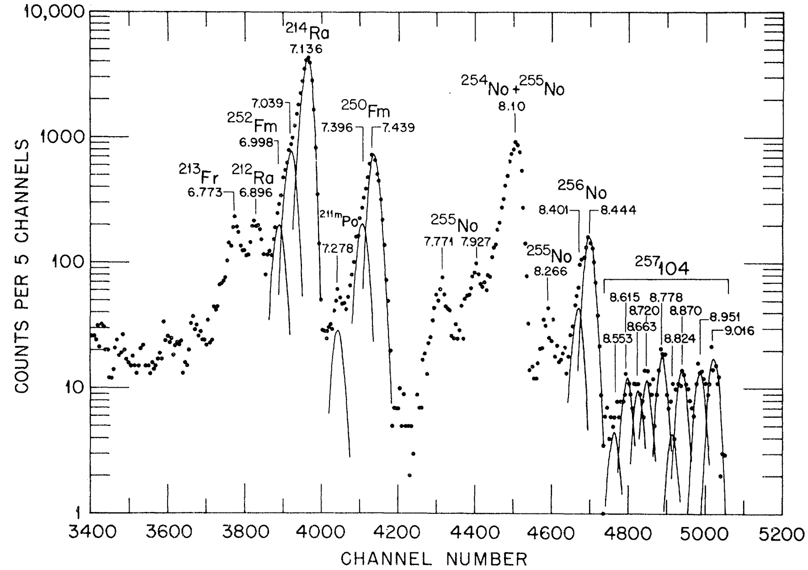

FIG. 1. α-particle energy spectrum for activities produced in the bombardment of ^{249}Cf with 73.0-MeV ^{12}C ions for a total of 31.3 μpA h. Counting time is 11 sec, and the energy range expected for α events from the decay of ^{257}104 is indicated.

energy to K-series x rays for $Z>100$) was ~900 eV FWHM. The absolute full-energy-peak detection efficiency for photons of this energy originating on the catcher disk was 14.9%.

The following information was stored for each detected α particle: (1) the α-particle pulse height, (2) the pulse height of any coincident photon, (3) the time between the detection of the initiating α particle and the coincident photon (in the range 0–100 μsec), (4) the time difference in milliseconds between the arrival of the disk in the counting assembly and the observation of the decay event, and (5) a counting-cycle sequence number. These five parameters were transferred to a SEL 840A computer using a fast on-line data-acquisition system,[14] where they were first stored on disks and later transferred to magnetic tape for permanent storage.

Some 30 000 10-sec counting cycles were performed during which approximately 3000 atoms of ^{257}104 were produced and about 1000 α particles from the decay of these atoms were observed. An α-particle energy spectrum representing a portion of our data is shown in Fig. 1. α particles arising from the decay of ^{257}104 are expected to lie in the energy range between 8.5 and 9.1 MeV.[11-13]

In Fig. 2, we show our experimental photon-energy spectrum in histogram form for the energy range 100 to 160 keV for those photons correlated in time ($0<t<100$ μsec) with all α particles in the energy range 8.5 to 9.1 MeV. The K-series x-ray spectra predicted for elements with $Z=100$, 101, 102, and 103 are also shown in Fig.

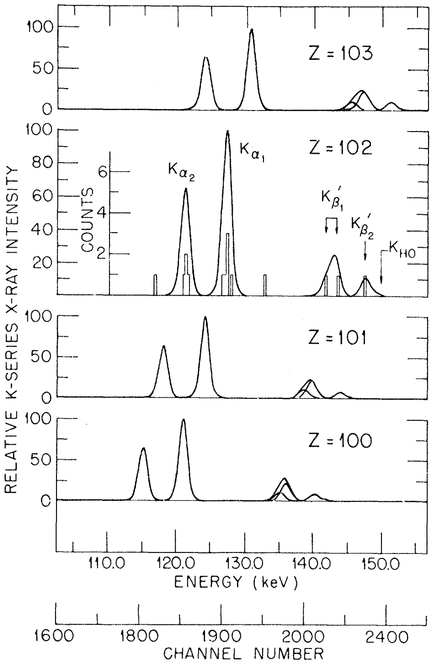

FIG. 2. Characteristic K-series x-ray spectra expected for the elements with $Z=100$ through 103. The experimental photon spectrum coincident with α particles in the energy range 8.5–9.1 MeV is shown in histogram fashion under the curve labeled 102 and forms the basis of a conclusive identification of element 104.

2. These curves were constructed using the calculated energies of Carlson *et al.*, the calculated intensities of Lu, Malik, and Carlson,[9] and include the effect of our instrumental energy resolution. Our measured energies for the $K\alpha_2$ and $K\alpha_1$ lines are 120.9 ± 0.3 and 127.2 ± 0.3 keV, respectively, and compare very favorably with those predicted by Carlson *et al.* for $Z = 102$: 121.020 and 127.42 keV, respectively.

The intense α group at 7.136 MeV due to ^{214}Ra (half-life measured in these experiments, 2.46 ± 0.03 sec) was used to assess the possible influence of random uncorrelated coincidence photons on the spectrum shown in Fig. 2. Since the ^{214}Ra α group proceeds to the ground state of ^{210}Rn, no real coincidence photons are possible. On the basis of this analysis, we expect 0.4 ± 0.1 random coincident photon events in the entire photon energy range as shown in Fig. 2 for the full 100-μsec time window. The delayed photon event at 133 keV was most probably a random coincident event; the photon recorded at an energy of 117 keV was a prompt coincident event associated with the 8.615-MeV α transition of ^{257}104 and is well accommodated in the energy-level sequence for ^{253}No proposed by us on the basis of this experiment.

It is quite clear from an examination of Fig. 2 that our experimental coincident photon spectrum can only match in energy and intensity the theoretical K x-ray spectrum for $Z = 102$. Therefore, since we have observed K-series x rays characteristic of atomic number $Z = 102$ in coincidence with α particles (with $Z = 2$), we have established that the atomic number of the α-decaying parent is $Z = 104$. This excellent agreement between calculated and observed K-series x-ray energies and intensities allows this technique to be applied to the short-lived heaviest elements which are produced on a one-atom-at-a-time basis. We have demonstrated that a conclusive atomic-number determination may be made with only a few thousand atoms.

Ten of the thirteen observed coincident K-series x rays were not prompt coincident events but arise from the internal conversion of a state in ^{253}No at ~300 keV with a half-life of 31.3 ± 4.1 μsec. A similar excited state occurs in other $N = 151$ isotopes.[15] The three prompt K-series x rays are associated with still higher-energy excited states in ^{253}No. Our measured K-series x-ray yield per decay of ^{257}104 is 0.16 ± 0.04, somewhat smaller than the value of 0.56 ± 0.04 observed for the isotope ^{255}No in our previous experiments[2,15] because of a different α-decay intensity pattern. A more complete description of the decay of the isotope ^{257}104, apart from the x-ray identification, is the subject of a publication currently in preparation.[16]

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Differential Energy Spectra of Low-Energy (<8.5 MeV per Nucleon) Heavy Cosmic Rays during Solar Quiet Times*

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Carbon, oxygen, and heavier nuclei have been observed below 8 MeV per nucleon during solar quiet times. We find that the C/O abundance ratio is 0.8 ± 0.4, the N/O ratio is 0.4 ± 0.25, and the differential energy spectra below 1 MeV per nucleon have the form $KE^{-4.9\pm0.3}$. We infer from this spectral form that most of these particles are likely to be of solar origin. The large errors on the abundance ratio do not allow a decisive answer to the likely origin.

Although extensive satellite and balloon measurements[1-5] have provided abundant information over most of the present solar cycle about the modulated galactic cosmic rays above roughly 15 MeV per nucleon, the extension of observations during solar quiet times below this energy has been limited to protons and α particles.[6,7] The continuous presence of low-energy protons and helium nuclei during solar quiet times was first established by Fan and co-workers[6,8] who also found unexpected upturns in the spectra below about 20 MeV and surprisingly little variation in the flux or spectral shapes during quiet periods from 1964 to 1967. Kinsey's[7] careful analysis of data from the Goddard dE/dx-versus-E experiments on IMP-3 and IMP-4, while revealing a continuous presence of protons and α particles down to 5 MeV per nucleon, showed substantial variability of these low-energy components over the period from May 1967 to August 1968. He concluded that during most of the time period particles below about 10 MeV per nucleon were substantially of solar origin.

In this Letter we present for the first time measurements of low-energy (<8 MeV per nucleon) heavy cosmic rays in interplanetary space during relatively quiet time periods. The data were obtained in October 1972 using a newly designed ultralow energy telescope (ULET) on board the IMP-7 (Explorer 47) satellite which was launched on 22 September 1972 into a nearly circular, 240 000-km apogee orbit. ULET operated successfully until 19 November 1972 when the thin window of the proportional counter ruptured. Because of this only a limited amount of quiet-time data could be collected. In this Letter we restrict ourselves to particles with a nuclear charge $Z \geq 6$.

The detector system[9] makes use of the dE/dx-versus-E method for particle identification and energy determination. To extend to 200 keV per nucleon the energy range over which two-parameter analysis can be made, we use a thin-window proportional counter (D_1) for the "dE/dx" measurement and a conventional, fully depleted, 700-μm-thick surface-barrier silicon detector (D_2) for the "E" determination. A plastic scintillator cylindrical cup anticoincidence detector S, which surrounds D_1 and D_2, is used to reject background and penetrating particles. The *total* thickness of material in front of the solid-state detector is 330 μg/cm^2, 140 μg/cm^2 of which is due to the isobutane counter gas. The geometrical factor of the telescope is 1.0 cm^2 sr. To obtain preferential an-